Sintering of Si_3N_4 Powder Prepared by Self-Propagating High-Temperature Synthesis (SHS)

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Abstract

Preparation processing of sintered silicon nitride ceramics was emphatically investigated with Self-Propagating High-Temperature Synthesis (SHS) of silicon nitride prepared by ourselves as raw material. The results indicate that good sinter ability can be obtained with cheaply SHS of silicon nitride preparing silicon nitride materials. The cost of silicon nitride materials will be lowered.

Keywords : self-propagating high-temperature synthesis (SHS), hot pressing, spark plasma sintering (SPS), Si_3N_4, mechanical properties

1. Introduction

Silicon nitride is a material well known among engineers for its high strength, toughness, and resistance to corrosion, low weight and ability to withstand high temperatures-all of which make it suitable for the toughest jobs[1,2].However,the relatively higher cost of ceramic components compared to metallic counterparts is a large obstacle that hampered its wider acceptance[3,4]. The high price of Si_3N_4 powder is an important factor leading to higher cost of silicon nitride materials.

It is well known that Self-Propagating High-Temperature Synthesis (SHS) method has many potential advantages such as low processing cost, energy-efficiency and high production rate[5]. If this Si_3N_4 powder will be applied to produce silicon nitride ceramics, the cost of Si_3N_4 productions will be lowered. In order to give full play to the advantages and potential of this material, the characters of β-Si_3N_4 powder synthesized by SHS by ourselves, the sintering process, the microstructure and the mechanical properties of Si_3N_4 ceramics with the addition of Al_2O_3 and Y_2O_3 as sintering additives are investigated in this study.

2. Experimental and Results

The SHS of β-Si_3N_4 powder (self-prepared) after acid washing and then mixed with 6.67 wt. % Y_2O_3 (purity >99 %, d_{50}=3.528 μm, Yuelong Chemical Industry, China) and 3.33 wt. % Al_2O_3 (purity >99%, d_{50}=2.553 μm, Yuelong Chemical Industry, China) was wet-milled for 2 h. After drying, powder mixtures obtained were sieved to 100 mesh., then dry-pressed uniaxially in a steel die to form cylindrical bodies of φ40 mm×10 mm at 30MPa. For comparison, hot-press (HP) sintering and spark plasma sintering (SPS) processes were used in this study.

The bulk density of the sintered compact, the bending strength, microhardness and the fracture toughness were measured.The morphology of the Si_3N_4 microstructure was characterized by scanning electron microscopy. Crystalline phases were identified by X-ray diffraction.

The results of the densification of the sintered samples were presented in Table 1.

Table 1. Density and relative density of the sintered samples

Sintering process	Sintering temperature (℃)	Ball-milled media	Density (g/cm^3)	Relative density (%)
HP	1650	distilled water	3.2188	98.37
HP	1650	alcohol	3.2385	98.97
HP	1700	distilled water	3.2537	99.43
HP	1750	distilled water	3.2122	98.16
HP	1750	alcohol	3.2151	98.25
HP	1800	distilled water	—	—
HP	1800	alcohol	—	—
SPS	1500	alcohol	3.2318	98.76

Morpholoy of the surface of the specimen was observed by SEM and shown in Fig. 1. The sample exhibited a microstructure with self-reinforcing rod-like β-Si_3N_4 grains forming an interlocking structure. The SEM fractograph of the sample was shown in Fig. 2.

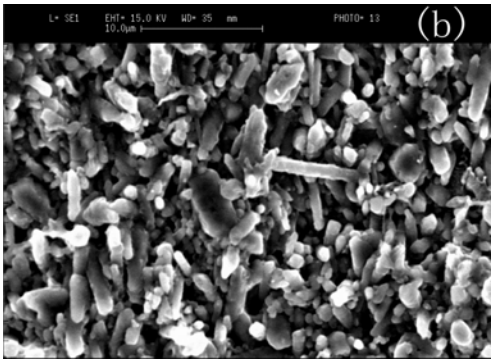

Fig. 1. SEM micrograph of the polished and etched surface of the specimen hot-pressed at 1700℃.

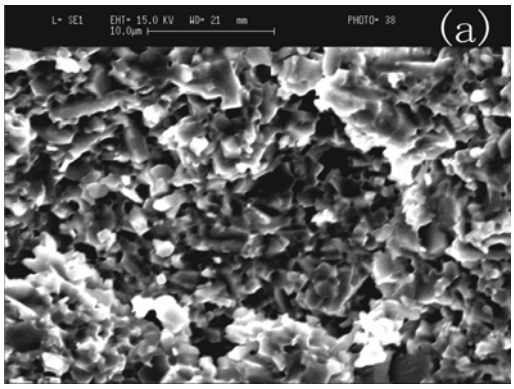

Fig. 2. Fractured surface of the specimen sintered by HP at 1700℃. Specimen were fractured at R.T.

The microhardness, three-point bending strength and the fracture toughness were resumed in Table 2.

Table 2. Mechanical properties of the sintered samples

Sinter-ing process	Sintering temperature (℃)	Micro-hardness H_V (GPa)	Bending strength (MPa)	Facture toughness (MPa · $m^{1/2}$)
HP	1650	14.76	623.88	4.98
HP	1650	14.94	654.71	4.34
HP	1700	16.73	611.72	5.72
HP	1750	15.58	466.09	5.85
HP	1750	15.42	445.54	5.79
SPS	1500	15.72	716.46	7.03

3. Summary

The results indicated that good sinter ability can be obtain ed with the cheaply SHS of silicon nitride powder preparing silicon nitride materials, which will make the cost of silico n nitride productions lowered. The results obtained were as f ollows:

1. The crystalline phases of Si_3N_4 powder used were mainly β-phase. The β-ratio of Si_3N_4, Rβ, is 97.6%. The morphology of rod-like grain with a diameter of 0.2~0.5μm and a length of 2~5μm.was observed.
2. The relative density of β-Si_3N_4 reached near to the full densification (99.43%) at the hot-pressing temperature of 1700℃, and the relative density of β-Si_3N_4 sintered by SPS at 1500℃ reached to 98.76%.
3. The samples tested all exhibited a similar microstructure with self-reinforcing rod-like β-Si_3N_4 grains forming an interlocking structure. A significant grain growth could be observed with increasing hot-pressing temperature. The microstructure of Si_3N_4 sintered by SPS at 1500℃ contained much finer grains than the microstructure of hot-pressed samples.
4. The better mechanical properties of hot-pressed Si3N4 were obtained at 1700℃.The hardness was16.73 GPa, toughness value was 5.72 MPa · m1/2 and the bending strength was 611.72 MPa. The mechanical properties of Si3N4 sintered by SPS technique at 1500℃were better than ones by HP. The microhardness was 15.72 GPa, its bending strength (716.46 MPa) and fracture toughness (7.03 MPa · m1/2) values were both highest in all results. It should be due the finer and homogeneous microstructure of Si_3N_4 sintered by SPS.
5. The effect of two ball-milled medias of distilled water and alcohol on the sinterability of this Si_3N_4 powder is not obvious.

4. References

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